



THE KOREA INDUSTRIAL
PROPERTY OFFICE

This is to certify that the annexed is a true copy from the original records
of the following application as filed with this office.

Application Number : 6514/1989

Date of Application : May. 18, 1989

Applicant(s) : Korea Research Institute of Chemical Technology

Sep. 14, 1990

COMMISSIONER

BEST AVAILABLE COPY

Applicant

Korean Research Institute of Chemical
Technology, President: Young Bok, Chae

Address; #100 Jangdong, Usung-ku, Daejonjikhalsi,
Korea.

Nationality; KOREA

Inventor

Name ; Tae Jin, BOM
Address ; 3-808 Dongsan Aptment Galma-dong
Seo-ku, Daejonjikhalsi, Korea.

Nationality; Korea

Inventor

Name ; Steven Say-kyoun OW
Address ; 21-142, Samboo Apartment, #407 Taepung-
dong, Joong-ku, Daejonjikhalsi, Korea.

Nationality; U.S.A

The Title of Invention

Biological Deinking Method

1. The title of the invention

Biological deinking method.

2. Detail description of the invention

This invention related to the process for reclaiming useful pulp fibers from wood containing or wood free wastepaper by a biological method in the deinking process.

Deinking of pulp fibers is essentially a laundering or cleaning process which the ink is considered to be the dirt.

Chemicals along with heat and mechanical energy, are used to dislodge the ink particles from fibers and to disperse them in the aqueous medium. The ink particles are then separated from the pulp fibers, either by washing or flotation or by using a modern hybrid process that combines the two elements.

The chemicals used for the conventional deinking process are surfactants of which functions are detergency to remove ink from fiber, dispersing action to keep the ink particles dispersed prevent redeposition on the fibers, and foaming action in the froth flotation of ink particles.

A typical surfactant is a long chain molecules with the hydrophobic part to the one end and the hydrophilic part to the other end. The hydrophobic part may be consisted of fatty acid, fatty alcohol, alkylphenols or other oil-soluble surfactants.

The hydrophilic part in the deinking surfactant usually consists of anion molecules such as carboxyl acid salts or sulfonic acid salts and nonionic molecules such as polyoxyethylenated chains.

The typical surfactants commonly used in the washing and froth flotation deinking processes are ; sodium and potassium salts of strait-chain fatty acid (soap), linear alkylbenzenesulfonate (LAS), - olefine sulfonate, long-chain fatty alcohol, polyoxyethylenated alkylphenols, alkylphenoethoxylates, and polyoxyethylenated strait-chain alcohols.

Major disadvantages of using these surfactants in the deinking process are excess foaming in the subsequent pulp stock flow and papermaking process lines. Some of the above surfactants are resistant to biodegradation in the effluent treatment stages causing a serious environmental problem.

In the froth flotation deinking process, a collector is added to agglomerate ink into large particles and attach them to the air bubbles. Collectors are required for effective flotation and are usually anionic long-chain fatty acid soap. Fatty acid collectors are precipitated with calcium ions to form larger, insoluble ink particles and collector particles. With injection of air in the flotation cells, the agglomerated ink particles adhere to the bubbles, rise to the surface and are skimmed off from the system.

Major disadvantages of the flotation method using the fatty acid collector is a pitch deposition and calcium scaling problems in the subsequent stock lines and papermaking process equipments.

Besides the surfactants, other chemicals are caustic soda, sodium silicate, metal ion chelating agents and hydrogen peroxide. The hydrogen peroxide bleaching agent has to be added in order to prevent a pulp color yellowing caused by the addition of caustic soda and to improve brightness of pulp fibers.

When an advance in the modern printing and photocopying technology the conventional deinking with the aid of surfactants encounters serious problems with the wastepaper printed with the use of heavily coated, highly polymerized, or nonimpact inks, such as ultraviolet, heatset, Xerox, laser and ink jet. These inks usually contain cured polymer resins which bind ink particles so strongly on the fiber surface that it is impossible to dislodge the inks completely during the wastepaper defiberizing (pulping) stage with the conventional deinking chemicals. Excess heat and mechanical energy are also required along with the ineffective conventional chemicals.

In the conventional floatation deinking process for newsprint wastepaper a major technical problem has something to do with the fine ink particles embedded in the fiber bundles and between fibrils which are almost impossible to be removed from the fibers by a washing and/or floatation process.

This invention provides a new and much improved deinking method which is effective in the newsprint deinking as well as the wood free printed wastepaper such as whiteledger, laser printed, xerographic copypaper and computer printout wastepaper.

This invented deinking method is to remove ink particles with the use of biological activity of enzyme on the cellulose fiber surface and a dispersing function of enzyme protein on ink particles.

In contrast to the conventional method no alkali and deinking surfactants are required although some surfactants can be used along with the enzyme to enhance the deinking efficiency. In the froth floatation process the fatty acid collectors are not required. Since caustic soda is not used in the newsprint deinking, hydrogen peroxide bleaching agent is not also required for the yellowing prevention.

The elimination of the fatty acid collector in this biological deinking process will solve the persistent pitch and scale deposition problem associated with the conventional floatation process using the fatty acid type soap and calcium salts and silicates.

The invented process is described in details as follows:

The newspaper such as old newsprint or printed wood free wastepaper is disintegrated in the conventional pulper (consistency 4-7%) or in the high consistency pulper, 12-15%, at the water temperature ranged from room temperature up to 80 C. The addition level of enzyme is 0.005% to 5.0% based on dry weight of wastepaper, pH of the stock slurry is adjusted in the range of 3.0 to 8.0. As compared to the conventional pulping process using caustic and surfactants the pulping in the process of enzyme can be completed in a relatively short period and ink particles are completely separated from the fiber surface and dispersed well. The dispersed inks are removed out of pulp fibers by the conventional washing process equipments such as vibration screen and drum washer without an aid of detergent surfactants in a single and multi stages. The ink particles dispersed with the action of enzyme protein can be also selectively removed out of the diluted pulp slurry with conventional floatation equipments which air is injected or drawn into pulp to provide bubbles to pick up the particles. No fatty acid

collector is required in the case of waste newsprint. But the small amount of fatty acid collector may be added to enhance the ink removal efficiency in the case of laser-printed wastepaper.

This biological deinking process is to lower pulping energy to a large extent since the addition of enzyme resulted in a substantial reduction in pulping time as compared to the pulping in the absence of enzyme, almost 50% reduction. The observed faster and easier pulping in the presence of enzyme may be attributed to a unique biological activity of enzyme which is effective to debond the fiber bonding and dislodge the inks bonded on the fiber surface as well as within the fiber bundles or between the fibrils. A partial enzymatic hydrolysis of cellulose within micro structure of fiber surface may occur during the pulping stage. Because of this biological activity of enzyme the fine ink particles embedded within fiber bundles, fibrils and fines which has been impossible to be taken out by the conventional deinking chemicals in the case of old newsprint deinking.

According to this biological deinking method of old newsprint, the addition of hydrogen peroxide to prevent the fiber yellowing is not required, which will result in a substantial reduction of deinking chemical cost as compared to the conventional deinking process using caustic soda, hydrogen peroxide, chelating agent and sodium silicates.

It should be pointed out that the physical strength properties of the resulting pulp fiber prepared by this invented method are found to be higher than those of the corresponding pulp prepared by the conventional method in addition to the much higher resulting pulp brightness. The enzyme addition does not appear to degrade the fiber strength, instead improve the fiber strength by not-yet unknown reasons.

Example 1.

Deinking of old newsprint with a cellulolytic enzyme.

A sample of old newsprint wastepaper was added to the pulper where was filled with 40 C water at the consistency of 4% and a cellulase was dissolved at the dosage level of 0.1% based on oven dry weight of wastepaper. The wastepaper was soaked for 10 minutes and then disintegrated for 5 minutes. ~~At this time~~ complete disintegration of wastepaper, the one half of pulp slurry was diluted to 1% consistency.

The diluted pulp slurry was moved to the air flotation cell and then the dispersed ink particles were removed out of the pulp slurry with skimming off the ink particles froth out of the cell while injecting air through a porous plate. The flotation time for the complete removal of the ink froth was one minute.

The other half of the pulp slurry was washed on a laboratory vibration screen to remove the dispersed ink particles.

The resulting recycled pulp fibers obtained by the flotation and the washing step were evaluated for the pulp brightness and the mechanical strength properties. To compare this enzyme-treated deinked pulp to the conventional deinked pulp, the same sample of wastepaper was treated in the pulper with addition of 1.0% NaOH, 0.3% H_2O_2 , 3% sodium silicate solution (water glass) and 0.8% of SBRFAX MT-90 (fatty acid soap) and 0.2% IGRPAL-660 based on oven dry weight of wastepaper. The pulping time was 10 minutes for a complete disintegration. After diluting to 1% consistency, the dispersed ink particles were removed by the flotation method with the laboratory flotation cell as the way described above.

As shown in Table 1, the brightness of the pulp deinked with enzyme was much higher than that of the pulp deinked with the conventional chemicals and the mechanical strength of the enzyme-deinked pulp was also superior to that pulp deinked with the fatty acid collector and the dispersant (IGRPAL-660). The microscopic observation revealed that the pulp prepared by the present invention contained more long fiber fractions and has smoother fiber surface and looks less mechanically damaged.

Table 1. Comparison of properties of recycled pulp by method of present invention and the conventional method.

		brightness (%)		tensile index (N.m/g)		tear index (mN.m /g)	
		KONP	AONP	KONP	AONP	KONP	AONP
present method	flotation	47.1	45.2	28.9	32.4	11.7	13.6
	washing	50.3	48.6	29.3	32.9	11.8	14.1
SBRFAX MT-90		45.1	38.4	30.1	32.8	10.8	13.1

KONP; Korean old newspaper.
AONP; American old newspaper.

The enzyme treated pulp gave cleaner and brighter pulp with the washing as compared to the flotation ink removal.

The enzyme addition appeared to accelerate the wastepaper disintegration to a large extent. When the old newspaper was disintegrated in the conventional pulper at the 4% consistency, the addition of 0.5% enzyme reduced the pulping time from 5 minutes (no enzyme addition) to 30 seconds for a complete disintegration as shown in Table 2.

Table 2. Relation between enzyme addition and disintegration time.

enzyme (%)	0.5	0.1	0
disintegration time (sec)	30>	60-120	300<

Example 2.

Deinking of laser CPO (computer printout) with cellulolytic enzyme.

It is almost impossible to achieve a complete removal of laser beam cured ink particles from the laser CPO wastepaper with the conventional deinking chemicals, because the ink particles are so strongly adhered to the fiber surface that alkali and general deinking surfactants in the conventional deinking chemicals are not able to dislodge and disperse in the pulp-water slurry.

A sample of laser-CPO wastepaper was added to the water in a laboratory high consistency pulper at the consistency of 12.6% and a cellulase was added to the water at the dosage level of 0.2% based on the dry weight of paper. At stock water temperature up 20-35 C, the pulping was carried out for 20 minutes. The completely disintegrated pulp slurry was diluted to 0.5% and then the dispersed ink particles was removed out of the pulp slurry using the laboratory flotation cell as the same way explained in Example 1. In this case, to increase the ink removal efficiency and selectivity a small amount of the conventional fatty acid collector, SERFAX MT-90, 0.3% based on dry weight of wastepaper was added prior to the air flotation and the flotation time was 3 minutes. To compare to the enzyme deinked pulp, the conventional deinked pulp was prepared by the same way but the different chemical conditions as follow:

1% NaOH on dry weight of wastepaper
 0.1% IGIPAL 660 dispersant
 0.8% SERFAX MT-90
 pulping temperature ; 60 C
 pulping time ; 30 minutes
 calcium salt addition to the flotation cell ; 200 ppm
 flotation time ; 3 minutes

The brightness and the strength properties of the resulting pulp samples were compared in Table 3.

As shown in the table, the image analysis of the paper samples indicates that the number of the residual ink particles was much less, about 10 times, for the pulp deinked with the enzyme and the tensile strength was also higher as compared to the pulp prepared with the conventional chemicals.

The recycled chemical pulp of high quality in terms of dirt count and fiber strength properties can be obtained with the use of enzyme in a combination of a small amount of fatty acid collector by the flotation method.



Table 3. Comparison of pulp properties recycled by the method of present invention and conventional method.

	brightness (%)	dirt amount (count/area)	tensile index (N.m/g)
enzyme+MT-90(0.3%)	79.0	450	34.3
MT-90 (90%)	80.6	4,330	26.3

Example 3.

Deinking of waste newsprint by pectinolytic enzyme.

As the same method to example 1, the waste newsprint containing 0.1% of pectinase was soaked for 10 minutes at 40 C and disintergrated for 5 minute. Diluting the disintergrated pulp to 1%, ink particles are removed by flotation for 1 minute.

As shown in Table 4, the brightness and the tensile strength of paper sheet prepared by method of present invention are improved.

Table 4. Comparison the method of using pectinolytic enzyme with conventional method.

	brightness (%)	tensile index (N.m/g)
present method	44.2	33.3
MT-90 (0.8%)	38.4	32.8

What we claimed is ;

1. Biological deinking method characterizing pulping of waste printed paper with enzyme and removing ink particles from fibers by flotation and/or washing method.
2. Biological deinking method characterizing using one kind of enzyme ~~with~~ ^{cellulase and/or pectinase,} in claim 1.
3. Biological deinking method characterizing adding the amount of enzyme in the range of 0.005% to 5% based on dry weight of wastepapers in claim 1 or 2.
4. Biological deinking method characterizing controlling the temperature of pulping process ranged from room temperature upto 60 C in claim 1.
5. Biological deinking method characterizing controlling the pH of pulping process ranged from 3 to 8 in claim 1.

대한민국 특허청

THE KOREA INDUSTRIAL
PROPERTY OFFICE

첨부의 등본은 아래 출원서의 원본과 상위
증명함.

is to certify that the annexed is a true copy from the original
of the following application as filed with this office.

출원 번호: 1989년 특허출원 제 6514 호
Application Number

출원 년 월 일: 1989년 5월 16일
Date of Application

출원 인: 재단법인 한국 과학 연구소
Applicant (s)

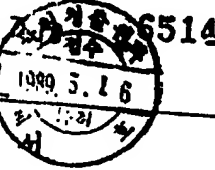
1990 년 9 월 14 일

특허청
COMMISSIONER



RECEIVED

1990 NOV 13 AM 7:27
GROUP 136



副本

출원

출원인	성명	재단법인 한국화학연구소 소장 제영복		국적	대한민국
	주소	대전직할시 용성구 장동 100번지			
대리인	성명	이돈상	변리사 등록번호	제 420 호	
	주소	서울특별시 강남구 역삼동 823-23		전화번호	557-2350
발명자	성명	오세근	주민등록번호	C696325	국적 미 국
	주소	대전직할시 중구 대평동 407번지 삼부아파트 21동 142호			
발명자	성명	엄매진	주민등록번호	570601-1674014	국적 대한민국
	주소	대전직할시 서구 갈매동 동산아파트 3동 608호			
	성명		주민등록번호		국적
	주소				

발명의 명칭	고체의 생분해성 발목에 의한 제생방법						
의 국 출 원	국명	출원외류	출원일자	출원번호	증명서류	우선권주장	
					첨부	미첨부	있음 없음

제 8 조의 규정에 의하여 위와 같이 출원합니다.

특허첨장 귀하. 1989년 5월 16일
대리인 이돈상
구비서류: 1. 출원서 정본 1통, 부분 2통 4. 외임장 1통
2. 출원심사청구서 1통
3. 명세서 정본 1통, 부분 2통

LEE-SANG, YIE
PATENT ATTORNEY

PHONE: 557-2350

명세서

1. 발명의 명칭

고지의 생물학적 발육에 의한 재생방법.

2. 발명의 상세한 설명

본 발명은 고지의 생물학적 발육에 의한 재생방법에 관한 것이다.

종래의 고지발육방법은 해리된 고지를 개면활성제 및 지방산 비누를 주성분으로 하는 발육제를 사용하여 선택법 또는 부상부유법에 의해 섬유로부터 분리된 잉크입자들을 제거시켜 왔다.

그러나 고지원료에 사용된 잉크의 구성성분 및 인쇄기술의 다양화 그리고 고지를 구성하고 있는 펄프의 종류가 다양해짐에 따라 발육제의 종류가 복잡해지고 있으며 발육방법에도 고도의 기술이 요구되고 있어 발육제의 종류와 공정이 복잡해지고 있다.

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☒ **BLACK BORDERS**
- ☒ **IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- ☒ **FADED TEXT OR DRAWING**
- ☒ **BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- ☒ **SKEWED/SLANTED IMAGES**
- ☐ **COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- ☐ **GRAY SCALE DOCUMENTS**
- ☒ **LINES OR MARKS ON ORIGINAL DOCUMENT**
- ☐ **REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- ☐ **OTHER:** _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.